TMI-2 CORE MATERIALS AND FISSION PRODUCT INVENTORY[a]

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ABSTRACT

Examinations have been performed to characterize the distribution of core materials and fission products at the damaged Three Mile Island Unit 2 (TMI-2) reactor. The purpose of this paper is to summarize the current status of the core materials and fission product inventory evaluations at TMI-2 and to present results that may affect reactor source term issues. Principal subjects include the relocation of core materials outside the reactor vessel, the formation of complex, core material interaction products, cesium retention in prior molten material, and the retention of tellurium, antimony, and ruthenium in association with core materials. Material examinations have now been performed on samples from throughout the reactor building and vessel. These examinations have helped define the end state core configuration and materials behavior. The examination results have been used to estimate the redistribution and inventories of core materials and fission products in the damaged reactor core for comparison with expected materials behavior, particularly for fission products.

INTRODUCTION

The Three Mile Island Unit 2 (TMI-2) pressurized water reactor underwent a prolonged loss of coolant accident on March 28, 1979, resulting in severe damage to the reactor core. As a consequence of the TMI-2 accident, numerous aspects of light water reactor (LWR) safety have been questioned, and the U. S. Nuclear Regulatory Commission (NRC) has embarked on a thorough review of reactor safety issues, particularly the causes and effects of severe core damage accidents. The nuclear community acknowledges the importance of examining TMI-2 in order to understand the nature of the core damage. Immediately after the accident, four organizations with interests in both plant recovery and acquisition of

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accident data formally agreed to cooperate in these areas. These organizations [General Public Utilities Nuclear Corporation (GPU Nuclear--owner/operator of TMI), Electric Power Research Institute (EPRI), the NRC, and the U. S. Department of Energy, collectively known as GEND] are presently involved in postaccident evaluations of TMI-2. The DOE is providing a portion of the funds for reactor recovery in those areas where accident recovery knowledge will be beneficial to the U. S. LWR industry. In addition, DOE is providing funds to acquire and examine samples obtained from the damaged reactor core.

The examinations of the TMI-2 reactor core began shortly after the accident with the evaluation of plant monitoring instruments[1], and sampling and analysis of the reactor coolant[2]. This was followed by examinations of samples of highly contaminated water, and sediment from the auxiliary and reactor buildings[3,4]. Much of this data has been analyzed and provides sufficient information to characterize the distribution of core materials outside the reactor core[5].

The end-state configuration of the TMI-2 core just after the accident as determined by closed circuit television (CCTV),[6,7] mechanical probing, and core boring operations[8] is illustrated in Figure 1. The initial examinations for core materials were performed on sections of control rod drive leadscrews, which extended from the upper plenum through to the beginning of the core proper[9,10]. Visual observations through the leadscrew guide tubes indicated the presence of a void region above a region of loose fuel debris (up to 94 cm deep) which was first sampled in October 1983[11,12]. As part of these examinations, it was determined that a crust layer was present below the debris bed. This crust layer was finally sampled using specialized core boring equipment adapted from the mining industry.

The visual examinations of the lower region of the reactor core (reference 8) indicated the presence of a central prior molten region contained by partly or fully metallic crust layers. Destructive examination of the core bores[13] obtained from the lower core indicate that the crust layers were composed principally of metallic constituents of the core that had not oxidized during the accident. Video examinations of the region below the reactor core were also performed, which indicated that prior molten material had relocated to the lower head of the reactor. Samples of this debris were obtained through the annulus between the thermal shield and the reactor vessel. Examination of this debris[14] indicated that it was a mixture of prior molten fuel, cladding, and structural materials which had oxidized and flowed down onto the lower head of the reactor. These examinations provide the bulk of the data on the distribution of core materials in the damaged reactor core.

The following sections discuss the distribution of the bulk debris in the reactor vessel, the segregation and redistribution of the elemental constituents of the various core components, and finally, a discussion of the redistribution of fission products and the possible effects on source term issues.

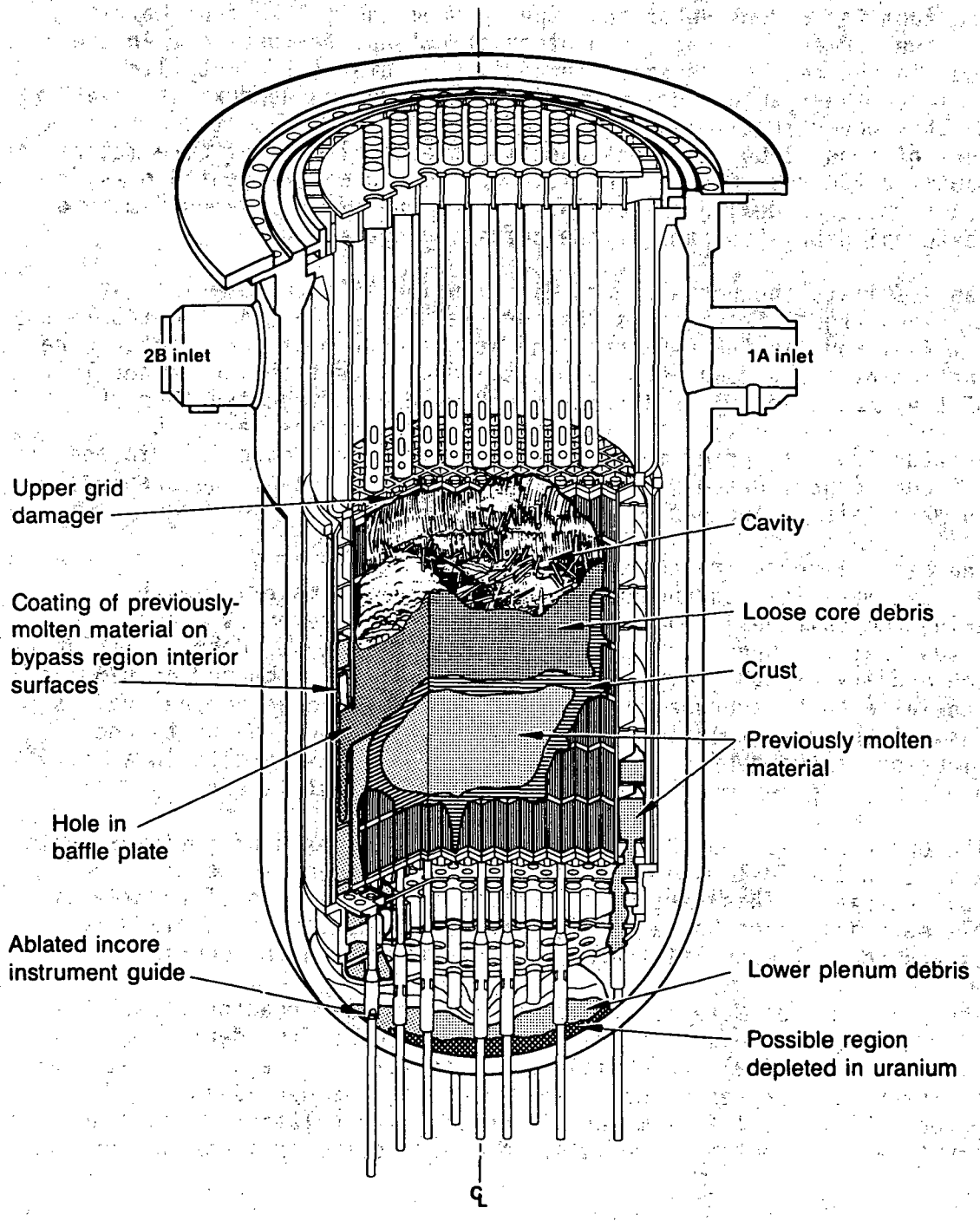

Figure 1. TMI-2 end state core condition. 8-9078

CORE MATERIALS INVENTORY

The original core materials inventory included approximately 94,000 kg of UO_2 fuel and 35,500 kg of cladding, structural, and control materials[15,16]. As a consequence of the accident, some material has been added to this inventory due to ablation of other reactor vessel materials and oxidation of some of the metallic structural components. Approximately 229 kg of structural steel was estimated to have been ablated from the underside of the upper plenum and 182 kg from baffle plates, the former core wall, and other structural features not in the core but in the reactor vessel. In addition, some of the metallic materials in the reactor core were oxidized producing approximately 459 kg H_2[17]. This would result in a total addition of oxygen as oxides to the core of about 3300 kg. Consequently, the total mass of core materials is about 133,250 kg. Of this total, about 100 kg was relocated outside the reactor vessel during the accident. GPU Nuclear has removed much of this external debris as part of the defueling operation.

Table 1 lists the postaccident distribution of core materials at TMI-2. The largest fraction of the core materials (33%) is located in the partial fuel rods in the periphery and at the bottom of the reactor core. Examination of these damaged assemblies indicate that they have not lost any of their core materials inventory and were not subjected to high temperatures. The remaining core material repositories range in composition from the prior molten fuel and structural materials in the central core consolidated region to the mixture of intact and previously molten materials in the upper core debris bed.

The central consolidated mass region is composed of a core of prior molten fuel materials surrounded by layers of crust material with differing compositions. Analysis of the data in references 8 and 13 indicates that the upper crust is composed of 2450 kg of debris with an average density of 8.3 g/cm^3, and the lower crust is composed of 8760 kg of material with an average density of 7.3 g/cm^3. These nominal values have associated uncertainties of 30-40% due to the heterogeneity and distribution of the debris in the crust layers. This results in a total of 25,990 kg of prior molten debris between the crust layers.

The upper debris bed (20% of core mass) is composed of a mixture of relatively intact fuel materials and prior molten fuel structural and control material. This part of the debris bed is made up of relatively friable material which has a density from 3-5 g/cm^3. Intact cladding shards and control material fragments were present in the debris.

The prior molten material relocated to the lower reactor vessel head (19,100 kg) has been examined only at the surface of the debris bed. This material (reference 19), which may not be representative of all material on the lower reactor vessel head, indicate that the debris is a homogeneous mixture of fuel materials (U,Zr) with relatively small amounts of structural and control materials. Nondestructive examinations of the

TABLE 1. ESTIMATED POSTACCIDENT CORE MATERIALS DISTRIBUTION

Core region	Estimated quantity(kg)	Uncertainty[a] (%)	Percent of Total core(%)
Intact fuel assemblies (Partially or fully intact)	44,500	5	33.4
Central core region resolidified mass	32,700	5	24.5
Upper core debris bed	26,600	5	19.9
Prior molten material on the Lower reactor vessel head	19,100	20	14.3
Lower core support assembly[b]	5,800	40	4.3
Upper core support assembly[b]	4,200	40	3.2
Outside the reactor vessel	100	-[c]	0.3

a. The uncertainty estimates are based on defueling. Those areas which have been defueled at this time have relatively low uncertainties[,] whereas those which have not have relatively high uncertainties.

b. The lower core support assembly is that portion of the reactor vessel below the core which includes the lower grid assembly and five flow distributor plates. The upper core support assembly is a coolant flow region outside the vertical baffle plates[,] which make up the peripheral boundary of the core.

c. Estimates of the amount of fuel material outside the reactor vessel are based on nondestructive evaluations of reactor components in the reactor and auxiliary buildings. They range from 60 to about 430 kg.

lower head suggest that structural materials may have relocated to this area of the reactor vessel[18], and examinations are in progress to better define the composition of this debris.

The remaining repositories in the reactor vessel (10,000 kg) have not been characterized, but are expected to be similar in composition to the material found on the surface of the debris bed on the lower reactor vessel head.

CORE MATERIAL DISTRIBUTION

Three principal types of core materials are present in the TMI-2 reactor core: the fuel rod constituents (uranium, zirconium, and tin), the control rod materials (silver, indium, and cadmium), and the structural materials (stainless steel and Inconel). Examinations by GPU Nuclear indicated that only about 100 kg of core material have relocated outside the reactor vessel, as previously discussed, and that the bulk of this material, a mixture of prior molten and intact fuel and structural materials, is located in the reactor coolant system in the steam generators and in the pressurizer. All other core materials remained in the reactor vessel.

Fuel Rod Constituents

Before the accident, the TMI-2 fuel rods consisted of 94,000 kg of UO_2, 24,000 kg of zirconium present mostly in the form of metallic Zircaloy, and approximately 370 kg of tin as a component of the Zircaloy. The UO_2, present as the oxide fuel, may be dissolved by the zirconium, which can be oxidized by steam as part of the high temperature melting process. During the accident at TMI-2, a significant fraction of the metallic zirconium was oxidized to ZrO_2 as indicated by the production of hydrogen. Approximately 43% of the Zr was oxidized during the accident; this estimate is based on the amount of H_2 (459 kg) produced by oxidation. However, some of the oxygen produced would have been used to oxidize structural metallic material (e.g., Fe, Cr). Consequently, the estimated 43% oxidation of Zr is conservatively high. Tin is expected to remain metallic in the conditions present during the accident at TMI-2[19], and its behavior during the accident would be expected to be different from the oxidized zirconium.

Analyses were performed on samples obtained from throughout the reactor core, which provided information on the redistribution of uranium in the reactor system. Table 2 lists the distribution of the principal fuel rod constituents in the various regions of the damaged reactor core. As indicated in the table, this distribution accounts for approximately 97% of the total uranium inventory. The principal repositories for uranium are the partial fuel assemblies, the upper core debris bed, debris relocated to the lower reactor vessel head, and the debris in the central consolidated region. The partial fuel assemblies around the periphery of the core (22.7%), and the partial assemblies below the central consolidated region (10.7%) make up the largest repository of core

TABLE 2. FUEL MATERIAL DISTRIBUTION IN THE REACTOR VESSEL

Core Material Repository	Core material distribution Percent of Inventory [a] Uranium	Zirconium	Tin
Upper reactor plenum	-b-	-b-	-b-
Upper core debris	24	13	-c-
Upper crust region			
- ceramic	1.3	1.2	2.3
metallic	--	0.3	6.1
Consolidated region			
ceramic	12	18	--
metallic	--	0.2	5.8
Lower crust region			
ceramic	3.6	2.8	9.3
metallic	--	5.6	26
Intact fuel rods	33	33	33
Lower reactor vessel head	15	11	-c-
Lower core support assembly	4.6	3.3	-c-
Upper core support assembly	3.3	2.4	-c-
Total	97	91	82

a. Percentage of the total amount of the element originally present in the core.
b. Insignificant amount (<0.1 wt%) based on the upper plenum measurements.
c. Elemental constituent not detected based on detection limits of approximately 0.1 wt%.

materials following the accident. Examinations indicate that the intact portion of these fuel rods show no evidence of accident damage.

The next largest repository for uranium at TMI-2 is the loose debris in the upper part of the reactor core. This material is particulate debris from 1 to 5 mm in diameter and is composed of prior molten fuel materials mixed with partial fuel pellets, cladding, and structural material pieces. This debris averages 75 wt% uranium which is higher than the core average concentration of about 66 wt%. The high uranium concentration is due to the melting and downward relocation of zirconium to the central part of the reactor core which is discussed in a following paragraph.

Approximately 15% of the core inventory of uranium was relocated to the lower reactor vessel head. Examinations performed on debris from the surface of the debris bed suggest that this material is a relatively homogeneous mixture of uranium and zirconium, with the uranium concentration (65 wt%) near the core average.

The consolidated core region, composed of the upper and lower crusts and the central core region, makes up the bulk of the remaining core inventory (17%). The uranium content is quite variable due to the high degree of heterogeneity of the materials in this part of the core. The average concentrations for uranium are: upper crust (49 wt%), central region (54 wt%), and lower crust (34 wt%), with a significant range of concentrations at all locations due to the heterogeneity of the debris. The lower crust contains only about half the expected amount of uranium as compared to the core average, indicating the presence of significantly more structural components in this part of the core. In general, these data suggest a very distinct segregation between the U-Zr-O and the metallic or oxidized structural materials. This is consistent with the expected behavior of uranium, in that it is expected to interact significantly only with the zirconium in the system. Structural materials are present as contaminants [and are] located in inclusions or at grain boundaries in the U-Zr-O matrix.

The zirconium data in Table 2 show a similar distribution to that indicated for uranium, except that the percentages of inventory are less, reflecting differences between the chemical behavior of the ceramic UO_2 and the metallic zirconium. The data indicate that almost half the zirconium originally present in the upper core (i.e., the upper debris bed) has relocated to lower regions of the reactor core. The data from the crust layers and the central core region indicate that the relocated zirconium contributed to the formation of the crust layers and was retained in the central core region as metallic inclusions. The high zirconium concentrations are present only in the metallic phase, and the data indicate that the zirconium forming these layers did not participate in the zirconium-water oxidation reaction and was transported as metallic zirconium or Zircaloy (melting point 2030 K) to form the layers in the central region of the core. This behavior suggests a core damage progression where the upper core region was not heated to high temperatures (>2200 K) until after the upper crust had formed.

The data for the debris relocated to the lower reactor vessel head indicate a lesser percentage of zirconium (about 4% of core inventory) for the quantity of uranium present. These data suggest that some zirconium did not remain with the uranium relocated to the lower reactor vessel head, but contributed to the formation of the lower crust. The summed data for the various repositories indicate that about 9% of the core inventory of zirconium has not been accounted for. Although there is a relatively large uncertainty due to the few number of samples examined relative to the mass of the reactor core, these data suggest the presence of additional Zr repositories.

The tin distribution in the reactor vessel is significantly different than that observed for the Zr. The core materials distribution data in Table 2 indicate that no significant amounts of Sn were found in the upper reactor plenum, the upper core debris bed, the debris on the lower reactor vessel head, and by inference from the lower reactor vessel debris data, in the lower and upper core support assemblies. The data indicate the presence of significant amounts (factors of 4 to 6 times greater than the core average concentration - 0.3 wt%) in the upper and lower crusts and the central core region. The data for the metallic region data indicate concentrations from 7 to 20 times the core average with higher concentrations in metallic inclusions in the central core region. The significant concentrations of Sn in the metallic region might be expected from the chemical behavior of Sn (i.e., a high free energy requirement for oxidation).

In the lower crust of the central consolidated region, there is evidence of accumulations of Sn in the metallic samples; however, in contrast to the upper crust there are also accumulations of Sn in the ceramic samples relative to Zr. These data suggest a scenario where much of the Sn in the upper part of the core flowed down and was either trapped as metallic inclusions in the ceramic melt or contributed to formation of the metallic lower crust.

The inventory data for tin in Table 2 suggest that approximately one third of the structural tin originally present in the core is now located in the lower crust and that almost half is present in the central core region.

Control materials

The TMI-2 reactor core contained 2200 kg of silver, 412 kg of indium, and 137 kg of cadmium control materials. These control materials were originally present in controls rods with 80% silver, 15% indium, and 5% cadmium. During the accident at TMI-2, a significant fraction of this material was expected to melt and possibly volatilize due to the relatively low melting point of the alloy (1072 K - 1123 K), and the low boiling points of the individual constituents. Analyses indicate that at TMI-2, control materials melted, and due to the pressure, did not produce a forcible ejection upon failure of the control rod cladding[20].

Table 3 lists the distribution of control material in the reactor vessel. The data indicate the presence of no measurable amounts of silver, the least volatile control material, on the lower reactor vessel

head, and by inference, in the upper and lower core support assembly regions. The data also indicate that a small amount of silver (10%) was deposited on the upper plenum surfaces. The bulk of this material was deposited with other fuel and structural materials as loose debris, which, based on particle size, could have been transported to the upper plenum as either a hydrosol or an aerosol. Examinations of samples from outside the core indicate that only small amounts of the control materials were released from the core, substantiating the analytical conclusion that a forcible ejection of the control materials probably did not occur.

Below the upper plenum structure is a void and the loose debris bed, which accounts for approximately 20% of the core mass. Based on the analytical results, only 1.8% of the core inventory of silver is contained in this mass. These data suggest that much of the silver (22%) melted and flowed down to form the crust layers and some was retained as inclusions in the central core region.

The silver data in Table 3 indicate that only about 47% of the core inventory of silver can be accounted for. Although the uncertainties associated with these data are relatively large due to the small percentage of the whole core examined, the data suggest additional repositories where a significant fraction of the silver inventory is located. The high density of metallic silver (10.5 g/cm^3--near the highest of the core materials) would suggest that it is deposited near the bottom of the core or possibly on the lower head of the reactor vessel. This is consistent with the lower crust data where the silver concentration (4.5 wt%) is the highest of any of the core regions examined.

Table 3 indicates that indium is concentrated in the central core region at higher relative concentrations than was the silver. The highest concentrations are in the lower crust at 1.1 wt%, which is 3-4 times the core average. This element is present in the metallic phases similar to silver. However, a significant amount is also present in the ceramic parts at concentrations 2-3 times the core average. If extrapolated to the mass of the various regions of the lower core, approximately 81% of the core inventory of indium is present in the lower core region. Although this estimate has a relatively large uncertainty, the data indicate that approximately 30% more indium than silver is located in the lower core, which suggests a different distribution of indium than silver and indicates that a significant fraction of the silver behaved differently than the indium and is located in another part of the reactor vessel[21].

The cadmium data indicate concentrations for the lower core that are similar or less than the core average concentration of 0.1 wt%. The data indicate lesser concentrations in the ceramic regions than the metallic regions, which might be expected as cadmium is not expected to oxidize. Much of the relatively volatile Cd was not released from the core but was retained in the central molten part of the core as a gas during the early part of the accident, probably as inclusions in the molten metallic material.

TABLE 3. CONTROL ROD MATERIALS DISTRIBUTION IN THE REACTOR VESSEL

Core material Repository	Core material distribution Percent of Inventory[a] Silver	Indium	Cadmium
Upper reactor plenum	1.0	-b-	-b-
Upper core debris	1.8	-c-	-c-
Upper crust region			
ceramic	1.2	3.6	0.65
metallic	2.4	3.3	0.39
Consolidated region			
ceramic	10	27	6.1
metallic	1.6	2.1	1.1
Lower crust region			
ceramic	7.3	7.2	1.4
metallic	11	16	2.9
Intact fuel rods[d]	11	11	11
Lower reactor vessel head	-c-	-c-	-c-
Lower core support assembly	-c-	-c-	-c-
Upper core support assembly	-c-	-c-	-c-
Total	47	70	23

a. Percentage of the total amount of the element originally present in the core.
b. Insignificant amount (<0.1 wt%) based on the upper plenum measurements.
c. Elemental constituent not detected based on detection limits of approximately 0.1 wt%.
d. Only 10.7% of the partially intact fuel assemblies contain control material as the balance (22.7%) are peripheral assemblies which do not contain control materials.

Structural materials

The distribution of structural materials in the various parts of the lower core region is discussed in detail in reference 22. The extrapolated data for the four elements considered (iron, nickel, chromium, and molybdenum) indicate extrapolated inventories up to 3 times the core inventory (only for molybdenum). The principal source of this bias is the central core region which has a non-representative sample distribution due to the core boring technique used to obtain the samples. If the central core region data are excluded, the inventories are below 100%.

The iron data for the reactor core indicate that the upper core debris bed is depleted in iron content relative to the percentage of core mass (i.e., 20%), but that there are significant concentrations of iron in the lower crust and the central, consolidated region. Iron makes up large percentages of the metallic phases in all regions of the lower core with the highest percentage in central region inclusions. Metallurgical data indicate that the iron is mostly (80%) not oxidized and is found alloyed with nickel and lesser amounts of chromium.

The bulk of the core inventory of chromium (>50%) is found in the lower core region at concentrations ranging from 1.4-1.7 wt% and that the bulk of this element is located in the metallic region. Metallurgical examinations of prior molten debris indicate similar concentrations to those found in the lower core, and that a fraction of the chromium is oxidized and retained in the grain boundaries of the prior molten fuel material.

Almost 50% of the core inventory of nickel is located in the lower crust. The data range from 2-6 times the core average concentration. The concentrations of Ni in the upper and lower crusts and the central consolidated region are similar and significantly higher than other portions of the reactor vessel. The Ni, as might be expected due to its high free energy requirement for oxidation, is located mostly in the metallic phases, with only a few percent located in the ceramic samples. The inventory data (i.e., 116% of core inventory) again indicate that the samples of the central core region are not representative of all the material in the central core, and are probably representative only of the fraction of the central core region that has a significant amount of metallic inclusions.

The molybdenum data indicate the bulk of the molybdenum is located in the metallic phases. As molybdenum is not a constituent of stainless steel, ratio comparisons were performed to evaluate the contribution of the Inconel core components (i.e., grid spacers, etc.) to the formation of the crust layers. The ratio data suggest that much of the metallic material that formed the upper and lower crust layers was relocated Inconel grid spacers. These data suggest that the formation of the crust layers in the core was controlled by the melt behavior of the grid spacers, and that other structural components contributed to a lesser extent to the formation of the crusts.

FISSION PRODUCT INVENTORY

The following sections discuss the distribution of fission products in the TMI-2 reactor system. This distribution is based on representative examples of each group of fission products that were measurable at the time the fission product measurements were made (4-7 years after the accident). The low volatility fission products include elements from the noble metals, some rare earths and actinides, tetravalents, and early transition elements. Generally, the oxides of these elements have low volatilities; however, some (e.g., LaO or CeO) have higher volatilities than do the elements. The only radionuclides from this group which were measurable during the lower vessel debris examination program were Cerium/Praseodymium-144, Europium-154, and Europium-155. The Ce-144 is produced principally by direct beta decay from fission, whereas the europium radionuclides are produced by neutron activation of fission produced species. The radionuclides produced by neutron activation have greater associated uncertainties with their data due to the neutron activation as it makes their production more dependant on neutron flux and consequently on the core location of the fuel material. The medium volatile species are Sr-90, Sb-125, and Ru-106. The strontium is expected to be an oxide whereas the antimony and ruthenium are expected to remain as metallics. The high volatiles are the noble gases, cesium, and iodine, which are released to a large extent at the temperatures found during a severe reactor accident.

Low Volatiles

Table 4 lists the fission product retention and distribution in the TMI-2 reactor system for the three low volatile fission products measured (i.e., Ce-144, Eu-154, and Eu-155). The low volatiles were not transported out of the reactor core and only a little fuel material was physically transported to the reactor coolant system. In the reactor vessel, the fuel material distribution is similar to the mass distribution. The Eu-154 and Eu-155 data is similar to that seen for the Ce-144, except for variations in the measured retention values. The relatively large uncertainty associated with the inventories of radionuclides (Eu-154) is due to the fact that this isotope is produced by neutron capture by another radionuclide. In this instance, Eu-155 is produced by neutron capture by Sm-154, a stable fission product. The data indicate that all low volatile fission products are retained in the reactor vessel.

Medium Volatiles

Table 5 lists the fission product distribution data for the medium volatiles. The data for Sr-90, the radionuclide in this category which exhibited the least mobility, indicates that only a small amount (<3.5 %) was released from the reactor vessel. The distribution is again similar to the fuel material distribution, which suggests that the bulk of the activity was retained in the prior molten fuel material. While the examination data indicate that the bulk of the Sr-90 is located in the ceramic parts of the samples, there has been some transport of the Sr-90 to the metallic region samples. Almost complete accountability is indicated for the Sr-90.

The Sb-125 data in Table 5 indicate high concentrations in the metallic portions of the lower reactor core. The metallic sample concentrations are 6-20 times those found in intact fuel. These data suggest that the Sb-125 remained in a metallic state. The percent of core inventory data indicate that 62% of the core inventory of Sb-125 was contained in the lower core.

The Ru-106 data listed in Table 5 follow a similar distribution to that observed for Sb-125, and indicate that much of the Ru-106 is found in association with metallic samples at concentrations 4-12 times those found in intact fuel. Of particular importance to the accident scenario is the timing of the transport of the metallic material to form the crusts and the metallic inclusions in the consolidated region. The metallic materials had to have been transported to form the upper regions of the core after the Ru-106 (boiling point 4423 K) and Sb-125 (boiling point 1653) were released from the fuel matrix.

High volatiles

Table 6 lists the distribution of the high volatiles (Kr-85, I-129 and Cs-137). These radionuclides were significantly relocated and transported to the reactor coolant, and consequently to the reactor building basement. The data for Kr-85 is accurate to within 5-10% as the quantity of intact fuel is known from defueling, and the measurement of this radionuclide in a known gas space (i.e., the reactor building) is also quite accurate. In addition to the approximately 85% accounted for in these measurements, it is expected that some fraction of the core inventory was also retained in the upper core debris bed.

The uncertainties associated with the I-129 data are significantly greater than the noble gases because I-129 is highly reactive and may plateout on surfaces, making accurate assessments difficult. Measurements of radioiodine concentrations in the reactor building basement indicate a wide range of concentrations in the particulate debris. Extrapolation of these concentrations to the quantity of debris present in the basement indicates that much of the core inventory of radioiodine remained in the reactor building basement. Table 6 shows the same retentions for iodine and cesium in the basement, as transport of the two radionuclides to the reactor building would be expected to be similar. However, a lesser fraction of the Cs-137 would be expected to plateout on surfaces; therefore much of the Cs-137 remained in solution in contrast to the iodine.

The Cs-137 data indicate almost complete retention of the cesium in the reactor building with only 5% released to the auxiliary building. This activity is located principally in the reactor coolant bleed tanks and the letdown demineralizers.

TABLE 4. LOW VOLATILE FISSION PRODUCT DISTRIBUTION IN THE REACTOR SYSTEM

Fission product Repositories	Fission product distribution Percent of Inventory [a]		
	Ce-144	Eu-154	Eu-155
Ex-vessel			
Containment atmosphere, basement, and tanks	.01	-b-	-b-
Reactor coolant system	-b-	-b-	-b-
Auxiliary building	-b-	-b-	-b-
In-vessel			
Upper reactor plenum	-b-	-b-	-b-
Upper core debris-A	26	30	24
-B[c]	20	19	19
Upper crust region	1.4	2.0	1.6
Consolidated region	24	32	22
Lower crust	5.9	7.9	5.1
Intact fuel rods	30	30	30
Upper core support assembly	3.4	4.5	-d-
Lower core support assembly	4.7	6.3	-d-
Lower head-reactor vessel	16	21	-d-
Total	105	122	110[d]

a. Percentage of the total amount of the fission product inventory calculated from comparisons with ORIGEN2.
b. Insignificant amount (<0.1 wt%) based on the measurement data.
c. Two sets of bulk sample measurements were performed on the upper debris bed. The A series was performed on samples from near the center of the core at a variety of depths whereas the B series were bulk samples from near the bottom of the debris bed. For the totals, the B series data were used.
d. Measurements not performed for this radionuclide at this core location. The total shown value in parenthesis is a total which assumes the same distribution as Eu-154 for the repositories where measurements were not performed.

TABLE 5. MEDIUM VOLATILE FISSION PRODUCT DISTRIBUTION IN THE REACTOR SYSTEM

Fission product Repositories	Fission product distribution Percent of Inventory [a]		
	Sr-90	Ru-106	Sb-125
Ex-vessel			
Containment atmosphere, basement, and tanks	2.1	0.5	0.7
Reactor coolant system	1	-b-	0.2
Auxiliary building	0.1	-b-	0.7
In-vessel			
Upper reactor plenum	-b-	-b-	-b-
Upper core debris-A	23	14	13
-B[c]	19	16	24
Upper crust region			
ceramic	0.73	0.8	0.5
metallic	-b-	3.8	7.8
Consolidated region			
ceramic	8.3	2.2	3.1
metallic	-b-	9.0	6.9
Lower crust			
ceramic	4.5	5.7	7.4
metallic	-b-	24	36
Intact fuel rods	30	30	30
Upper core support assembly	3.9	0.23	0.22
Lower core support assembly	5.3	0.32	0.30
Lower head-reactor vessel	18	1.1	1.0
Total	93	94	119

a. Percentage of the total amount of the fission product originally present calculated from comparisons with ORIGEN2.
b. Insignificant amount (<0.1 wt%) based on the upper plenum measurements.
c. Two sets of bulk sample measurements were performed on the upper debris bed. The A series was performed on 16 cm^3 samples from near the center of the core at a variety of depths whereas the B series were bulk samples from near the bottom of the debris bed. The data provide a range. For the totals, the B series data were used.

TABLE 6. HIGH VOLATILE FISSION PRODUCT DISTRIBUTION IN THE REACTOR SYSTEM

Fission product Repositories	Fission product distribution Percent of Inventory [a]		
	Cs-137	I-129	Kr-85
Ex-vessel			
Containment atmosphere,	-b-	-b-	54
basement water	47	(47)[c]	-b-
Reactor coolant system	3	1	-b-
Auxiliary building	5	7	-b-
In-vessel			
Upper reactor plenum	-b-	-b-	-b-
Upper core debris-A	5.3	5.9	-b-
-B[d]	4.3	5.3	-b-
Upper crust region	0.41	0.27	-b-
Consolidated region	0.77	2.1	-b-
Lower crust	1.4	3.5	-b-
Intact fuel rods	30	30	30
Upper core support assembly	0.46	0.12	-b-
Lower core support assembly	0.63	0.16	-b-
Lower head-reactor vessel	2.1	0.54	-b-
Total	95	103	85

a. Percentage of the total amount of the fission product originally present calculated from comparisons with ORIGEN2.
b. Insignificant amount (<0.1 wt%) based on the measurement data.
c. Wide range of concentrations and quantity of debris would indicate percentage of total inventory that could be greater than the core inventory. Consequently, the Cs-137 inventory is considered representative of the quantity of iodine deposited in the reactor building.
d. Two sets of bulk sample measurements were performed on the upper debris bed. The A series was performed on 16 cm^3 samples from near the center of the core at a variety of depths whereas the B series were bulk samples from near the bottom of the debris bed. The data provide a range. For the totals, the B series data were used.

In the reactor vessel, the data indicate similar distributions for both the Cs-137 and I-129. The data for the upper core debris indicates that about 20% of the original inventories of both Cs-137 and I-129 were retained in the debris. In the lower parts of the core, evidence suggests that some of the I-129 is associated with the metallic regions of the core. These data suggest that there are reaction mechanisms which cause accumulation of I-129 in the metallic sections, and indicate the probable chemical form of the I-129. The fraction of core inventory found in association with the lower core region prior molten material indicates the presence of only 2.4% of the core inventory.

The Cs-137 data in Table 6 indicate retentions ranging from 2.7 to 13% of the original content in the fuel material. Again, some of the activity is found in association with metallic regions, which provides information on the possible chemical form of the Cs-137, and indicates the presence of reaction mechanisms which define the behavior of Cs-137. The total inventory of Cs-137 present in the lower core is quite low at about 2.6% and indicates significant release and transport of Cs-137 from this part of the reactor core.

CONCLUSIONS

This section summarizes the results of the core material and fission product inventory analysis. The results are based on the examination of a relatively small number of samples obtained from the the TMI-2 reactor vessel. This analysis provides a reasonable accountability for greater than 95% of the core materials and greater than 90% of all fission products. Listed below are some of the specific observations or conclusions of this analysis:

- The distribution of the principal fuel rod constituents accounts for approximately 97% of the total uranium inventory and suggests that this distribution is relatively accurate
- Principal repositories for uranium are the partial fuel assemblies, the upper core debris bed, debris relocated to the lower reactor vessel head, and the debris in the central consolidated region
- Metallurgical data indicate a very distinct segregation between the uranium ceramic and the metallic or oxidized structural materials which is consistent with the expected behavior of uranium in that it interacts significantly only with the zirconium in the system
- Almost half the zirconium originally present in the upper core (i.e., the upper debris bed) has relocated to lower regions of the reactor core
- Relocated zirconium contributed to the formation of the crust layers and was retained in the central core region

- High zirconium concentrations are present only in the metallic phase and the data indicate that the zirconium forming these layers did not participate in the zirconium-water oxidation reaction and was transported as metallic zirconium to form the metallic layers in the central part of the core

- Much of the Sn in the upper part of the core flowed down and was either trapped as metallic inclusions in the ceramic melt or formed the lower crust

- The high density of metallic silver (10.5 g/cm^3--near the highest of the core materials) is consistent with the lower crust data, where the silver concentration (4.5 wt%) is the highest of any of the core regions examined

- The cadmium data suggest that much of this relatively volatile element was not released from the core but was retained in the central molten part of the core as a gas, and was deposited with other, probably liquid metallic constituents as inclusions in the cooling metallic material

- Only a small fraction of low volatile radionuclides and fuel material were physically transported to the reactor coolant system

- In the reactor vessel the fuel material distribution is similar to the mass distribution

- The distribution of Sr-90 is similar to the fuel material distribution[,] which suggests that the bulk of the activity was retained in the prior molten fuel material

- The percent of core inventory data indicate that much of the core inventory of Sb-125 could be contained in the lower core with the bulk of the inventory located in the central core region

- The Ru-106 data listed in Table 5 follow a similar distribution to that observed for the Sb-125 data[,] and indicates that much of the Ru-106 is found in association with metallic samples at concentrations 4-12 times those found in intact fuel

- The Kr-85 data indicate accountability of 85% of the core inventory with other probable repositories in the reactor core

- The Cs-137 data indicate almost complete retention of the cesium in the reactor building with only 5% released to the auxiliary building. This activity is located principally in the reactor coolant bleed tanks and the letdown demineralizers.

REFERENCES

1. H. Lawrowski, A Report on Transport of Radioactivity from the TMI-2 Core to the Environs, Report to the President's Commission on the Accident at Three Mile Island, August 29, 1979.

2. S. Langer et. al., "Fission Product Release Pathways in TMI-2," Proceedings of the ANS TMI-2 Topical Meeting on the TMI-2 Accident, October 31 - November 4, 1988.

3. C. A. Pelletier et. al., Preliminary Radioiodine Source Term and Inventory Assessment for TMI-2, GEND-028, March 1983.

4. C. V. McIsaac and D. G. Keefer, TMI-2 Reactor Building Source Term Measurements: Surfaces and Basement Water and Sediment, GEND-042, October 1984.

5. S. Langer et. al., TMI-2 Fission Product Inventory Program, FY-85 Status Report, GEND-057, November 1986.

6. Quick Look Inspection: Report on the Insertion of a Camera into the TMI-2 Reactor Vessel Through a Leadscrew Opening, GEND-030, Vol. 1, March 1983.

7. J. Adams and R. Smith, Lower Plenum Video Data Survey, EGG-TMI-7429, July 1987.

8. E. L. Tolman et. al., TMI-2 Core Bore Acquisition Summary Report, EGG-TMI-7385 Rev. 1, February 1987.

9. K Vinjamuri et. al., Examination of H8 and B8 Leadscrews from Three Mile Island Unit 2, GEND-INF 052, September 1985.

10. G. M. Bain and G. O. Hayner, Initial Examination of the Surface Layer of a 9-Inch Leadscrew Section Removed from Three Mile Island-2, EPRI NP-3407, January 1984.

11. D. W. Akers et. al., TMI-2 Core Debris Grab Samples--Examination and Analysis, GEND-INF-075, September 1986.

12. G. O. Hayner, TMI-2 H8A Core Debris Sample Examination, GEND-INF-060 Vol 2, May 1985.

13. D. W. Akers et. al., The TMI-2 Lower Core Region: Examination and Analysis, GEND-INF-092, November 1988.

14. C. S. Olsen et. al., Examination of Debris from the Lower Head of the TMI-2 Reactor, GEND-INF-084, January 1988.

15. G. R. Eidam et. al., "TMI-2 Defueling Conditions and Summary of Research Findings," IAEA/NEA International Symposium on Severe Accidents in Nuclear Power Plants, Sorrento Italy, March 21-25, 1988.

16. TMI-2 Accident Core Heat-Up Analysis - A Supplement, NSAC-25, June 1981.

17. J. O. Henrie and A. K. Postma, Analysis of the TMI Unit 2 Hydrogen Burn, GEND-INF-023, Vol IV, Rockwell International, March 1983.

18. Analysis of Gamma Scanning of In-Core Detector #18 (L-11) in Lower Reactor Vessel Head, GPU Nuclear Corporation TPO/TMI-175, June 1985.

19. D. Cubicciotti and B. R. Singh, "Vaporization of Core Materials in Postulated Severe Light Water Reactor Accidents," Nuclear Technology, Vol. 67, November 1984.

20. D. A. Petti, Silver-Indium-Cadmium Control Rod Behavior and Aerosol Formation in Severe Reactor Accidents, NUREG/CR-4876, April 1987.

21. M. Hansen, Constitution of Binary Alloys, New York: McGraw-Hill Book Company, 1958.

22. D. W. Akers and R. K. McCardell, "Core Materials Relocation and Behavior in the TMI-2 Reactor Vessel," Proceedings of the ANS Topical meeting on the TMI-2 Accident, October 31 -November 4, 1988.